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**WO 01/74946 A1**

(54) Title: USE OF DENDRIMERS AS A PROCESSING AID AND SURFACE MODIFIER FOR THERMOPLASTIC RESINS

(57) Abstract: Dendrimer additives incorporated in thermoplastic resins (such as polycarbonate resins) are effective to alter the surface characteristics of the thermoplastic resin. Such compositions can be injection molded to produce articles in which the dendrimer additive is concentrated at the surface of the article to alter the properties of the resin. By selection of the type of dendrimer additive, the resulting characteristics of the molded article may be controlled.

**USE OF DENDRIMERS AS A PROCESSING AID AND SURFACE MODIFIER  
FOR THERMOPLASTIC RESINS**

**BACKGROUND OF THE INVENTION**

This application relates to the use of dendrimers as a component of thermoplastic resin compositions, such as polycarbonates, to achieve desirable surface properties in molded articles.

Polycarbonate is widely used as an engineering thermoplastic owing to its unique combination of toughness, stiffness, high softening-temperature and processability. However, molded articles formed from polycarbonates have several limitations such as low scratch, abrasion and chemical resistance and susceptibility to UV degradation. To counter these limitations, coating have been applied to the surface of the molded article. For example, scratch resistant coatings applied to molded polycarbonate allows it to be used as optical and headlamp lenses. UV-protective coatings allow polycarbonate to be used in exterior applications. Coatings are also applied for decorative and aesthetic purposes. For example, molded polycarbonate articles can be coated to look like chrome, wood, painted metal and the like. Such coated articles can have a soft, luxurious feel, and provide reduced glare. One the other hand, coating are typically applied in a separate step, which reduces productivity and adds to the cost of the product. Moreover, most coating are applied by a solvent evaporation technique which creates solvent emissions and requires a solvent recovery system. This further adds to the cost of manufacturing.

The alternative to applying a coating to a molded article is to incorporate an additive within the bulk of the polycarbonate resin that provides the desired functional characteristics. For example, siloxane copolymers may be added to improve chemical resistance, while antistatic agents are added to minimize static charge build-up. Conductive carbon black has been utilized as an additive to facilitate electrostatic painting of the molded article. Unfortunately, while these additives are beneficial in

the specific property for which they are added, they usually alter other material properties in an undesirable fashion. In particular, such additives frequently make the polycarbonate more brittle, and more susceptible to molding defects. In addition, they may be associated with a reduction in low-temperature ductility, softening temperature, modulus and increased plate-out.

US Patent No. 5,820,808 describes a method for making surface-modified articles using small amounts of an additive which is incompatible with the bulk polymer. The additive is dispersed within the bulk polymer as discrete droplets , and because of its incompatibility is driven to the surface in the injection molding apparatus. It has been reported, however, that adding incompatible additives to a polymer resin tends to result in delamination in injection molded articles. (Bucknall, C.B., *Toughened Plastics*, Applied Science, London (1977). Moreover, the addition of incompatible additives tends to reduce physical properties of the final molded article.

Thus, there remains room for improvement in the production of molded articles with desirable surface properties.

#### SUMMARY OF THE INVENTION

The present invention provides compositions comprising:

- (a) a thermoplastic resin; and
- (b) a dendrimer additive in an amount effective to alter the surface characteristics of the thermoplastic resin. These compositions can be injection molded to produce articles in which the dendrimer additive is concentrated at the surface of the article to alter the properties of the resin. By selection of the type of dendrimer additive, the resulting characteristics of the molded article may be controlled.

#### BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 shows the structure of a highly-branched hydroxy-terminated dendritic polymer of a polyester type;

FIGS. 2A-C show ToF-SIMS spectra for dendrimer alone (FIG. 2A), the surface of a disk molded from polycarbonate plus dendrimer (FIG. 2B), and the 5 spectra of the surface exposed by removing a 100  $\mu\text{m}$  layer (FIG. 2C); and

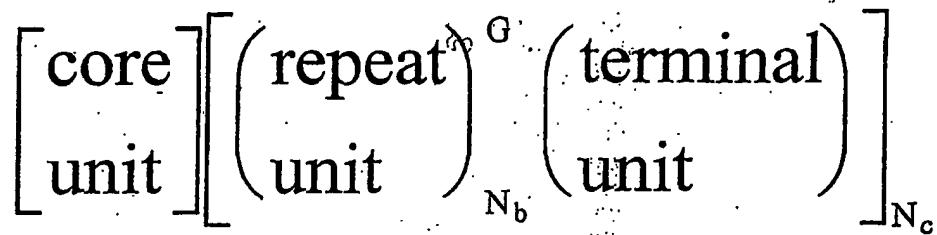
FIG. 3 shows the test set-up used to measure ductility of molded optical disk substrates.

#### DETAILED DESCRIPTION OF THE INVENTION

The present invention provides thermoplastic resin compositions with improved characteristics. The compositions comprise a thermoplastic resin and a 10 dendrimer additive component.

As used in the specification and claims of this application, the term thermoplastic resin refers to thermoplastic polymers and copolymers in general, and to blends thereof. A preferred thermoplastic resin is polycarbonate, which may be used in any of the various grades known in the art, including optical grades and high 15 molecular weight resins used for making impact-resistant articles. Other non-limiting examples of thermoplastic resins which may be suitably employed in the compositions of the invention are polyethylene terephthalate, polybutylene terephthalate, polyphenylene ether, polyamide, polypropylene, blends of polyphenylene ether and polystyrene, blends of polyphenylene ether and polyamide, 20 polyetherimide and blends of polyetherimide and polypropylene.

As used in the specification and claims of this application, the term “dendrimer additive” refers to dendritic and hyperbranched polymers. Such polymers have a densely branched structure. Dendrimers with identical repeat unit can be characterized with the formula



Although the present application is not limited to the use of such dendrimers, this formula is useful for characterization of the meaning of the term "dendrimer" in the specification and claims of this application. Thus, a dendrimer following this general form has a central core unit and some number of branches ( $N_c$ ) attached to the core. For purposes of this invention, the number  $N_c \geq 3$ . Fig. 1 shows the structure of a highly-branched hydroxy-terminated dendritic polymer of a polyester type suitable for use in the invention for which  $N_c = 3$ . Within each of the  $N_c$  branches attached to the core there are subbranches leading to terminal units. This is represented in the general formula by the terms  $G$ , which represents the number of generations of dendrimer synthesis and  $N_b$  which is the branch multiplicity of the repeat unit. For purposes of the present invention, both  $G$  and  $N_b$  have a value of  $\geq 2$ .

Dendrimer additives used in the invention may be made using a variety of polymeric backbone materials, and with terminal groups consistent with the properties to be obtained. Examples of specific backbone groups which may be employed include polyesters, polysiloxanes, polyolefins, polystyrene and polyamides. Examples of specific terminal groups hydroxy groups, alkyl groups, epoxy groups, silanol, acrylate and methacrylate esters, trialkoxysilane, unsaturated fatty esters, aliphatic and aromatic carboxylic acid esters and tertiary amines. Materials for use as dendrimer additives in the compositions of the invention are available commercially, and their synthesis is known in the art, for example from US Patents Nos. 5,777,129 and 6,037,444 and the patents and publications cited therein.

The terminal groups of the dendrimer additives are selected to achieve the surface properties which are desired in the final molded article. For example, incorporation of hydroxy-terminated dendrimers increases the static surface charge on molded articles making them suitable for use in electrostatic painting. Conversely, 5 methyl-terminated dendrimers reduce the static surface charge on molded articles. Thus, methyl-terminated dendrimers might be suitably employed in compositions for optical applications. Other end groups can be used to achieve other surface properties, including the following:

	<u>End Group</u>	<u>Surface Property</u>
10	Hydroxyl	Static
	Alkyl	Reduced Static, moisture resistance
	Acrylate	Chemical and Scratch Resistance
	Epoxy resistance	Chemical Resistance, Hydrolytic
15	Halogenated hydrocarbon, siloxane, phosphate esters	Flame retardancy
	Benzophenone, benzotriazole, amines, phenyl salicylate	UV Resistance
20	Low surface energy groups such as fluorinated hydrocarbons	Stain Resistance

In addition to specific modifications of the surface characteristics of molded articles, it has also been found that incorporation of dendrimer additives results in an increase in the melt flow index (MFI) of the composition. This increase is indicative of an improvement in the processability of the polymer. Thus, the dendrimer additives of the invention provide improved properties beyond any enhanced surface characteristics, and these improvements in processability are independent of the nature of the terminal groups of the dendrimer.

The dendrimer additives are incorporated in the compositions of the invention in an amount effective to alter the surface characteristics of the thermoplastic resin after molding. Because the dendrimer additives tend to migrate to the surface of a molded article, the amount of dendrimer additive employed can be quite low. Thus, in general, the amount of dendrimer additive will be in the range of from 0.1 to 5% by weight of the composition.

In addition to the thermoplastic resin and the dendrimer additive, the compositions of the invention may include additional additives known in the art for modifying the properties of a thermoplastic resins. Thus, the compositions may include impact modifiers such as styrene-butadiene thermoplastic elastomers, fire-retardant additives, colorants, thermal stabilizers, antioxidants, antistatic agents and flow promoters.

While not intending to be bound by a particular mechanism, it is believed that the mechanism of action through which dendrimer additives migrate to the surface of molded articles is different from that described in US Patent No. 5,820,808, because it does not depend on incompatibility between the dendrimer additive and thermoplastic resin. Rather, the differential migration arises as a result of rheological difference between the dendrimer additive and the thermoplastic resin. When a thermoplastic resin with a low concentration of dendrimer is subjected to a non-uniform deformation flow field, the resin chains (because of their substantially linear shape) migrate away from regions of higher deformation. Conversely, the dendrimers, which have a

unique somewhat spherical shape, migrate towards regions of higher deformation. The driving force for this differential migration is entropic in nature. The thermoplastic resin chains are more ordered in the region of higher deformation and hence have lower entropy. These chains therefore migrate towards regions of lower 5 deformation, where they can achieve more random conformations, with higher conformational entropies. On the other hand, the dendrimers, due to their extremely high mobility (diffusion coefficient), small size and low viscosity move into the region of higher deformation which has been vacated by the thermoplastic resin. The extent of such deformation-induced migration depends on the rate of deformation, and 10 the molecular characteristics of the thermoplastic polymer and the dendrimer, such as relaxation time and translational diffusivity. (See Agarwal et al., "Migration of macromolecules under flow: the physical origin and engineering implications", *Chem. Eng. Sci.* 49 (11): 1693-1717 (1994); Bhave et al., "Kinetic theory and rheology of dilute non-homogeneous polymer solutions", *J. Chem. Phys.* 95(4): 2988-3000 (1991) 15 ).

The practical effect of the migration of thermoplastic resins to regions of lower deformation and of the dendrimer additive to regions of higher deformation can be seen from an analysis of an injection molded product. An injection-moldable composition is formed by mixing a thermoplastic resin (such as polycarbonate) with a 20 small concentration of dendrimer additive. When the injection molding mixture is forced into the cavity during the mold-filling process, the material undergoes large deformation rates (for example 1,000 to 100,000 sec<sup>-1</sup>). Particularly, the material experience large elongational gradients near the advancing flow front and large shear rates and rate gradients near the mold wall. The combination of large 25 shear/elongational rates and their gradients results in the migration of dendrimer towards the walls of the mold. As the material reaches the wall, it is cooled down rapidly, thereby producing a surface which has a higher concentration of dendrimer than the bulk. At the same time, the core of the molded article has a depleted concentration of dendrimer, such that desirable properties of the bulk resin are

minimally effected. This distribution of dendrimer additive was confirmed using Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS) to evaluate a molded optical grade polycarbonate disk fabricated with methyl-terminated dendrimer. As shown in Figs. 2A-C, the peaks associated with the dendrimer alone are found to a significant extent in the surface portion of the disk, but not in the bulk polymer. Thus, when the compositions of the invention are used to prepare molded articles, the articles have a central bulk region and a peripheral surface region, with the majority (i.e., in excess of 50 %) of the dendrimer additive being located in the surface region.

The compositions of the invention may be utilized to form a variety of molded articles consistent with the applications in which the included thermoplastic resins are normally used. Specific, non-limiting examples of types of molded articles include optical disks, optical lenses, building and construction components, water bottles, electronic equipment housings, office equipment, electrical appliances, health care components and cellular phone housings.

The invention will now be further described with reference to the following non-limiting examples.

#### EXAMPLE 1

Polycarbonate (PC) blends were prepared from 98 to 100 parts of a polycarbonate resin having  $M_w$  of more than 18,000 with 0.1 to 2 parts of a dendritic polymer. The dendritic polymers used were obtained from Perstorp Specialty Chemicals, Sweden, and are sold with the product designations of H30, H3200 and E1. Characteristics of these products are summarized in Table 1.

The blends were prepared by mixing the constituents in pellet, powder or liquid form (as supplied by the manufacturer) and extruding them in a twin-screw extruder. The extruder was operated at 300 rpm with the temperature of the metering zone at 270 °C. Optical disk substrates (diameter = 120 mm, thickness = 1.2 mm) were injection molded using the extruded composition. Values for melt flow index

(MFI) were determined at 250 °C under 1.2 kg load to provide a quantitative measure of the processability of the compositions.

The ductility of the molded substrates was measured in terms of energy absorbed during a compression-bending test in a specially designed jig fixture. A 5 schematic representation of the test set-up is shown in Fig. 3. A disk was placed between the jigs at an initial separation of 105 mm. The lower jig is moved upwards at 500 mm/min, and the energy expended to bend the disk is monitored. The maximum stroke was 95 mm.

10 Static charge on the molded disks, immediately after molding, was monitored using a static charge meter.

Table 2 summarizes the compositions tested and the results in the aforementioned tests. It can be seen that MFI of all of the PC-dendrimer blends is high than that of the PC resin. Increasing amounts of dendrimer increased the MFI. Furthermore, the ductility of the disks was found to be the same for the blends and the 15 unblended PC. Thus, PC-dendrimer blends offer improved processability (as reflected in the increased MFI) without adverse effects on the mechanical properties of the molded disks. Furthermore, a dramatic reduction in static values was observed for blends containing an inert dendrimer (H3200) with alkyl (methyl) terminal groups. Increased static values were observed for blends containing the hydroxylated 20 dendrimer (H30).

## EXAMPLE 2

A disk prepared using the H3200 dendrimer at a level of 0.1 % was evaluated by ToF-SIMS to assess the distribution of the dendrimer within the disk. ToF-SIMS uses a pulsed ion beam to remove molecules from the very outermost surface of a 25 sample (about 10 nm), and can be performed at a sufficiently low intensity to insure that molecules are not broken into constituent pieces. Spectra as shown in Figs. 2A-C were obtained for neat dendrimer H3200 (Fig. 2A), the surface of the disk after

molding (Fig. 2B) and the surface of the disk after removal of the layer approximately 100  $\mu\text{m}$  thick from the surface of the molded disk. (Fig. 2C). The arrows in Figs. 2A and 2B indicate characteristic peaks associated with the dendrimer H3200. These peaks clearly appear in the spectrum of the disk, as molded, but are essentially absent from the spectrum for the disk with the surface layer removed. Thus, the dendrimers are preferentially located within the top 100  $\mu\text{m}$  of the surface of the disk. This allows dendrimers to be used to manipulate the surface characteristics of the molded article while minimizing the effect on the bulk properties of the base resin.

**Table I:** Molecular characteristics of dendritic polymers

Dendrimer	End-group	Number of End-groups/molecule	Molecular weight
H30	Hydroxy	32	3500
H3200	Methyl	32	3500
E1	Epoxy	11	10500

5

**Table II:** Flow (MFI), Ductility (Energy), and Static data for a few representative PC-dendrimer blends

	Composition						
	A	B	C	D	E	F	G
PC (MW=18,242)							
H3200	100	100	100	100	100	100	100
H30	0	0.1	0.5	0	0	0	0
E1	0	0	0	0	0	0.1	0.5

MFI (cc/10 min)	12.7	13.1	14.2	13.0	13.9	12.9	13.9
Energy (N.m)	5.01	5.03	5.01	5.11	5.08	5.16	5.08
Static (kV)	-3.10	-0.81	-0.07	-6.4	-8.6	--	--

## WHAT IS CLAIMED IS:

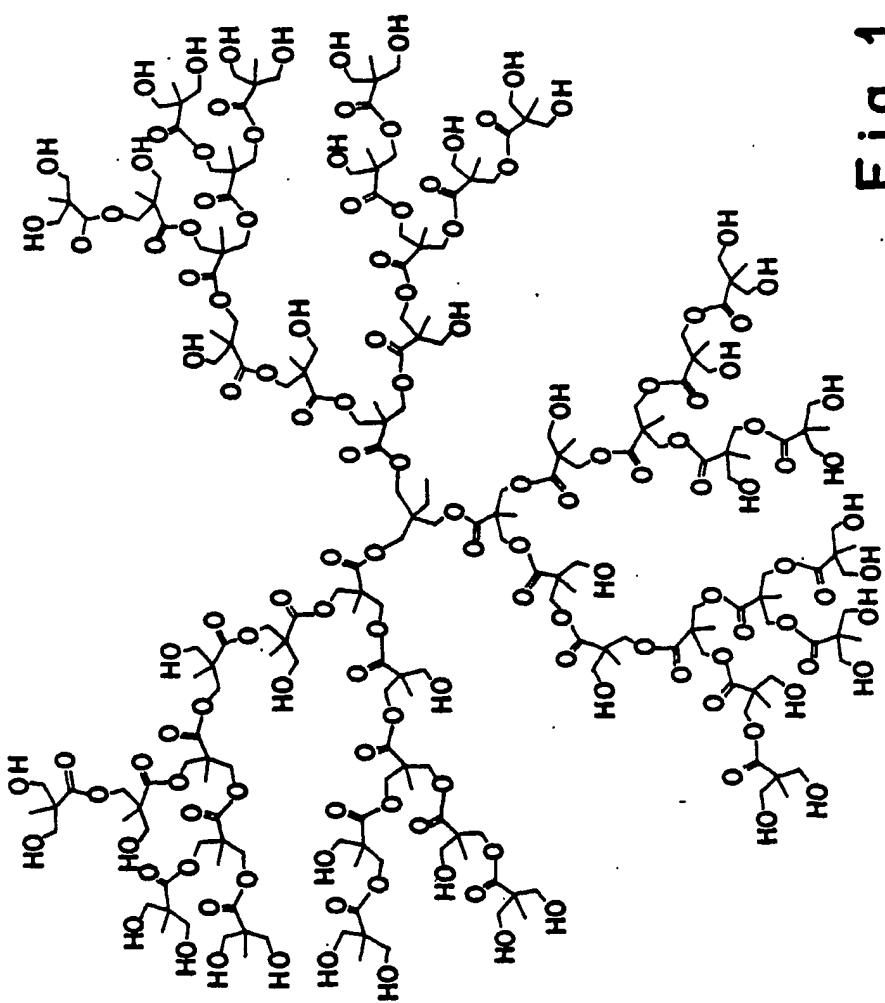
1. A composition comprising:
  - (a) a thermoplastic resin; and
  - (b) a dendrimer additive in an amount effective to alter the surface characteristics of the thermoplastic resin.
- 5 2. The composition of claim 1, wherein the dendrimer additive is present in an amount from 0.1 to 5% by weight of the total composition.
3. The composition of claim 2, wherein the thermoplastic resin is a polycarbonate resin.
4. The composition of claim 3, wherein the dendrimer additive is present in an 10 amount from 0.1 to 5% by weight of the total composition.
5. The composition of claim 1, wherein the dendrimer has terminal groups selected from the group consisting of hydroxyl, alkyl, acrylate, epoxy, halogenated hydrocarbon, siloxane, phosphate esters, benzophenone, benzotriazole, amines, phenyl salicylate, and fluorinated hydrocarbons.
- 15 6. A molded article formed from a composition comprising:
  - (a) a thermoplastic resin; and
  - (b) a dendrimer additive in an amount effective to alter the surface characteristics of the thermoplastic resin, wherein the molded article has a central region and a surrounding surface region, and wherein the majority of the dendrimer additive is located within the surrounding surface region.
- 20 7. The molded article of claim 6, wherein at least 50 % of the dendrimer additive is located within the surrounding surface region.

8. The molded article of claim 7, wherein the dendrimer additive is present in an amount from 0.1 to 5% by weight of the total composition.
9. The molded article of claim 7, wherein the thermoplastic resin is a polycarbonate resin.  
5
10. The molded article of claim 9, wherein the dendrimer additive is present in an amount from 0.1 to 5% by weight of the total composition.
11. The molded article of claim 6, wherein the dendrimer additive is present in an amount from 0.1 to 5% by weight of the total composition.  
10
12. The molded article of claim 6, wherein the thermoplastic resin is a polycarbonate resin.  
13. The molded article of claim 12, wherein the dendrimer additive is present in an amount from 0.1 to 5% by weight of the total composition.  
14. The molded article of claim 6, wherein the article is an optical disk, the thermoplastic resin is an optical grade polycarbonate, and the dendrimer additive is a dendrimer having alkyl terminal groups.  
15
15. The molded article of claim 6, wherein the dendrimer has terminal groups selected from the group consisting of hydroxyl, alkyl, acrylate, epoxy, halogenated hydrocarbon, siloxane, phosphate esters, benozphenone, benzotriazole; amines, phenyl salicylate, and fluorinated hydrocarbons.  
20
16. A method for modifying the surface properties of an article formed by injection molding a thermoplastic resin composition, comprising the step of adding to the thermoplastic resin composition prior to injection molding a dendrimer additive in an amount effective to alter the surface characteristics of the thermoplastic resin.

17. The method of claim 16, wherein the dendrimer has terminal groups selected from the group consisting of hydroxyl, alkyl, acrylate, epoxy, halogenated hydrocarbon, siloxane, phosphate esters, benozphenone, benzotriazole, amines, phenyl salicylate, and fluorinated hydrocarbons.
- 5 18. The method of claim 16, wherein the dendrimer additive is added in an amount from 0.1 to 5% by weight of the thermoplastic resin composition.
19. The method of claim 18, wherein the dendrimer additive comprises hydroxyl terminal groups, whereby the static charge on the surface of the article are increased.
- 10 20. The method of claim 16, wherein the dendrimer additive comprises alkyl terminal groups, whereby the static charge on the surface of the article are decreased.
21. A method for making a molded article comprising the steps of:
  - (a) preparing a composition comprising a thermoplastic resin, and a dendrimer additive in an amount effective to alter the surface characteristics of the thermoplastic resin; and
  - 15 (b) introducing the composition into a mold shaped to form the article, wherein the process of introducing the composition into the mold deforms the material with a large rate of deformation sufficient to produce a molded article having a central region and a surrounding surface region, and wherein the majority of the dendrimer additive is located within the surrounding surface region..
- 20 22. The method of claim 21, wherein the dendrimer additive is present in an amount from 0.1 to 5% by weight of the total composition.

23. The method of claim 22 wherein the thermoplastic resin is a polycarbonate resin.
24. The method of claim 23, wherein the dendrimer additive is present in an amount from 0.1 to 5% by weight of the total composition.
- 5 25. The method of claim 21, wherein the dendrimer has terminal groups selected from the group consisting of hydroxyl, alkyl, acrylate, epoxy, halogenated hydrocarbon, siloxane, phosphate esters, benozphenone, benzotriazole, amines, phenyl salicylate, and fluorinated hydrocarbons.

Fig. 1



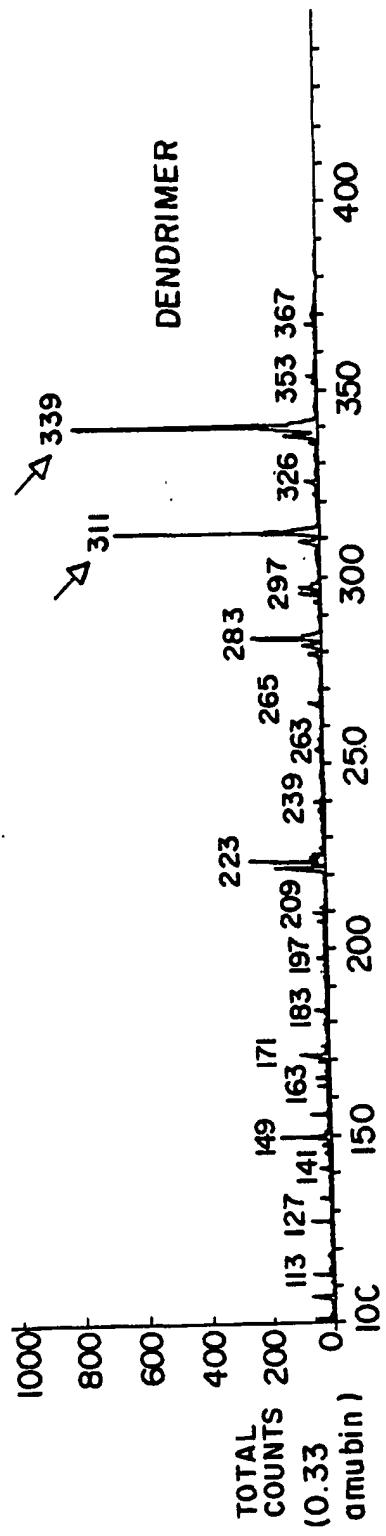


Fig. 2A

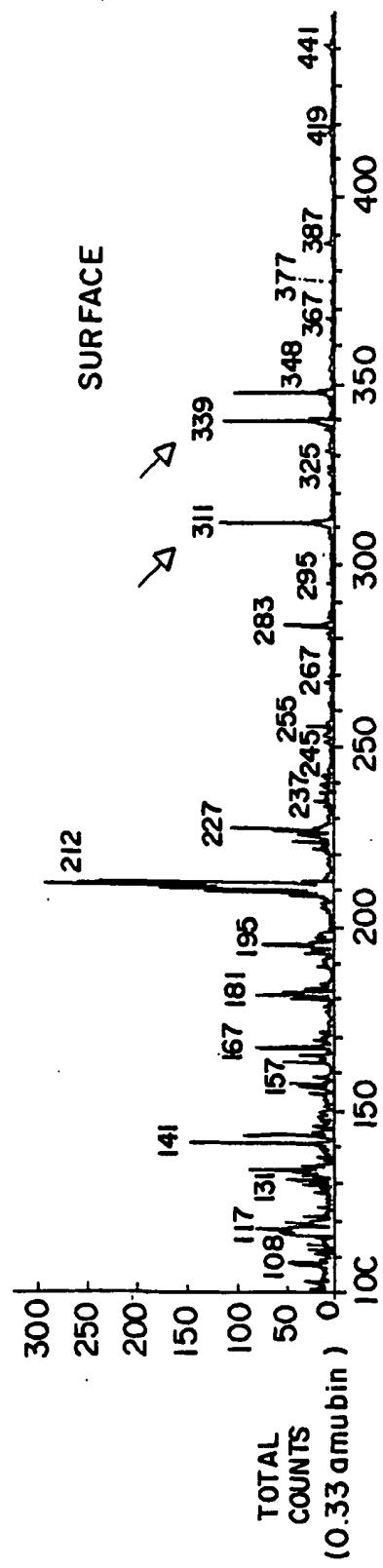


Fig. 2B

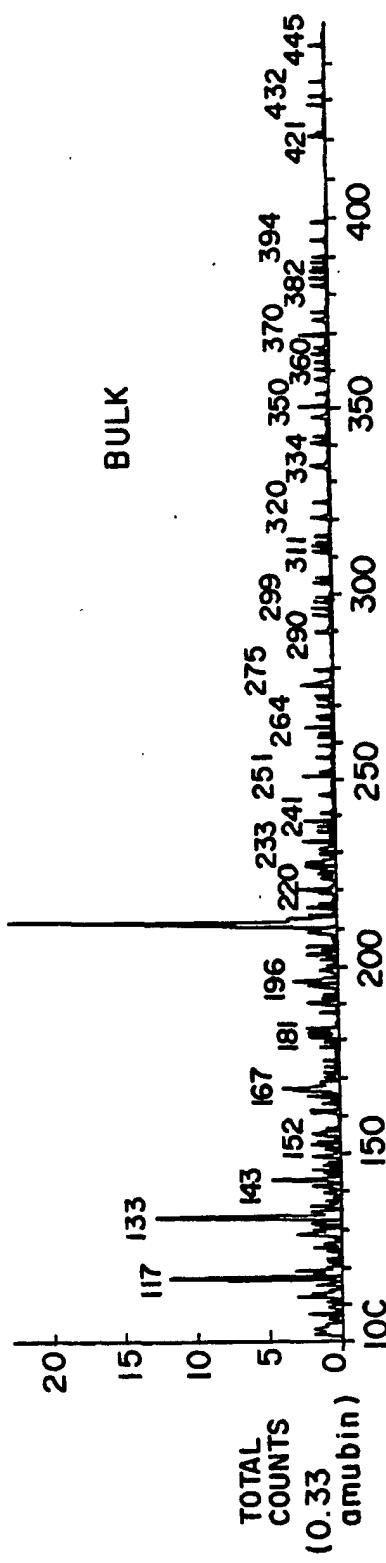


Fig. 2C

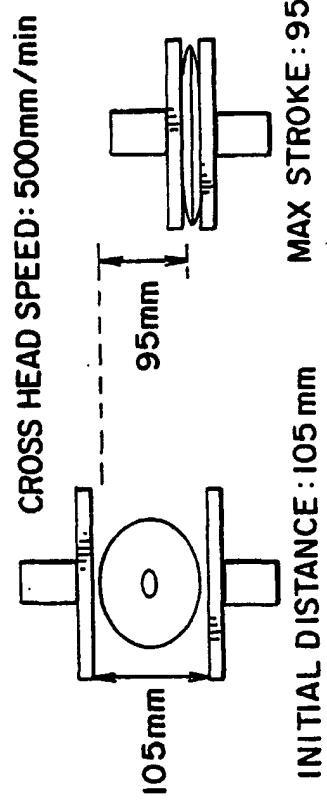


Fig. 3

## INTERNATIONAL SEARCH REPORT

onal Application No

PCT/US 01/40014

A. CLASSIFICATION OF SUBJECT MATTER  
 IPC 7 C08L69/00 B29C45/00 //C08L101:00

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
 IPC 7 C08L B29C C08G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, PAJ, EPO-Internal

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

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 Further documents are listed in the continuation of box C. Patent family members are listed in annex.

## ° Special categories of cited documents :

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- \*&\* document member of the same patent family

Date of the actual completion of the international search

Date of mailing of the International search report

25 June 2001

06/07/2001

## Name and mailing address of the ISA

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## INTERNATIONAL SEARCH REPORT

... Application No  
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## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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